

KINETICS OF THE ACCUMULATION OF STORED ENERGY IN THE
CRYSTALS OF ALKALI HALIDES IRRADIATED WITH PROTONSYe. K. Zavadovskaya, A. V. Kuz'mina,
Ye. M. Golovchanskiy, V. G. Vakhromeyev

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KINETICS OF THE ACCUMULATION OF STORED ENERGY IN THE
CRYSTALS OF ALKALI HALIDES IRRADIATED WITH PROTONSYe.K. Zavadovskaya, A. V. Kuz'mina,
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ABSTRACT. The authors study the kinetics of accumulation of stored energy in NaCl, KCl, KBr, and KI crystals after irradiation at room temperature with 4.5-MeV protons. The intensity of irradiation was 4.2×10^{10} protons/sec \times cm², varying from 15 min to 6 hr. Samples 5 x 5 x 4.5 mm were used.

Irradiation of solids results in an accumulation of point and complex defects in them. Separation of the effect of either type of defects upon the physical properties of a solid is possible by studying the kinetics of accumulation of the stored energy along with the study of the changes of physical properties within a wide range of radiation doses. At low doses of irradiation, the point defects will be of paramount importance in affecting the properties of the crystals. As the doses and intensity of irradiation increase, the point effects become converted to aggregates, correspondingly changing the physical properties of the crystals.

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Study of the kinetics of accumulation of stored energy and of the changes in optical properties and microhardness following gamma irradiation has shown that the extent of these changes in alkali halide crystals depends upon the type of the crystal, namely, for the crystals of LiF, NaCl, KCl, KBr, and KI the amount of stored energy, concentration of color centers and hardening is that much greater the greater is the lattice energy of the crystals [1,2,3]. In addition, it has been shown that under identical conditions the main contribution to stored energy and change of physical properties in the crystals with lesser lattice energy is supplied by complex defects, which cannot be determined by optical methods [3]. These studies were conducted within a small interval of irradiation doses from 1×10^8 roentgen to 5×10^8 roentgen and only for four values of irradiation doses. It seems of interest to follow the kinetics of the changes in stored energy and crystalline properties within a considerably wider range of irradiation doses and in greater detail.

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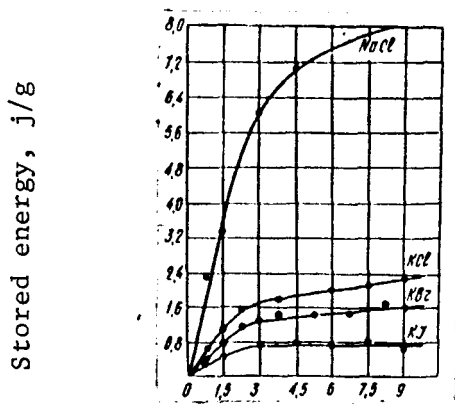
In order to conduct such a study it is essential that an irradiation source is selected which would afford a broad interval of irradiation doses within a comparatively short span of time (hours) and which would allow variation of the irradiation intensity. Such requirements are fulfilled by a cyclotron.

*Numbers given in the margin indicate pagination in original foreign text.

This work investigates the kinetics of accumulation of stored energy in NaCl, KCl, KBr, and KI crystals after irradiation at room temperature with protons having energy of 4.5 Mev using the cyclotron of Tomsk Polytechnic Institute. The intensity of the irradiation was 4.2×10^{10} protons/sec \times cm². The time of irradiation varied from 15 minutes to 6 hours, which provides an integral irradiation dose from 3.78×10^{13} to 9.7×10^{14} protons/cm². As the depth of penetration of protons of this energy does not exceed 250 microns, the irradiation of the samples was conducted on both sides. The dimensions of the samples were as follows: 5 x 5 x 4.5 mm.

The energy measurement was performed by dissolving the samples in an automatic microcalorimeter apparatus constructed at the Tomsk Polytechnic Institute [4].

Figure 1 illustrates the kinetics of accumulation of the energy stored in NaCl, KCl, KBr, and KI crystals as a function of the exposure dose of proton irradiation. The plot indicates that the magnitude of the stored energy for all irradiation doses increases with increased lattice energy of the crystal. As the irradiation dose increases, the kinetics of accumulation of stored energy for all the crystals has two clearly defined stages: a rapid growth of the stored energy up to an irradiation dose of 3×10^{14} protons/cm², which is followed by a slow linear growth. With decreasing crystal lattice energy the growth rate of the stored energy decreases. In the case of KI crystals, a saturation value for the stored energy is observed.



irradiation dose, 10^4 protons/cm²

Figure 1. Kinetics of Accumulation of Energy Stored in NaCl, KCl, KBr, and KI Crystals as a Function of the Exposure Dose of the Proton Irradiation.

This characteristic of the relationship between the stored energy and the dose of proton irradiation may have the following possible explanation. During the first stage the main contribution to the stored energy is that of F-centers, generated by irradiation. As the concentration of F-centers increases, their radiation and time burn-off becomes more intense, on one hand, and on the other, the aggregation of F-centers into complex electron centers is more effective. The first of these processes leads to the establishment of a certain thermodynamic equilibrium between the generated and collapsed F-centers and then leads to saturation, while the second process would contribute to the stored energy, but to a lesser extent than during the generation of individual F-centers. Let us clarify

this process. According to recent presentations, the complex electron centers represent aggregates of F-centers: M-center is 2F centers, R-center is 3F centers, etc. [5]. Contribution to the stored energy of one M-center is greater than that of two F-centers. As long as the concentration of M-centers and other complex centers is small, it is possible to neglect their contribution to the

stored energy. However, as soon as the concentration of M-centers becomes commensurate with that of the F-centers, one can no longer ignore the contribution of the complex color centers to the stored energy and the accumulation curves of the stored energy again show a more rapid linear growth with increased irradiation doses.

In order to justify the above assumptions one is required to measure the kinetics of accumulation of electron color centers in the investigated crystals under analogous irradiation conditions. However, regrettably, it is impossible to do since within the investigated interval of the irradiation doses the crystal become very intensively colored. After only 15 to 30 minutes of irradiation the color becomes so dense that it cannot be measured using spectrophotometer SF-4. The study of the kinetics of the accumulation of stored energy along with the kinetics of accumulation of color centers will be reported on in a subsequent presentation. Two methods shall be employed for this purpose. The first one, developed at the Tomsk Polytechnic Institute by D. I. Vaysburd [6,7], consists of the following procedure. By decreasing the energy of protons by means of aluminum foil it is possible to decrease the depth of penetration of protons into the alkali halide crystals to only few tens of microns. At this thickness of the colored layer it is possible to use an SF-4 spectrophotometer to measure the color density in hundreds and thousands of units per 1 cm, which corresponds to the concentration of defects of the order of 5×10^{18} to 10^{19} cm^{-3} and above. In using this method an assumption was made that the effectiveness of the formation of the color centers is independent of the energy of protons.

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The second method will permit broadening of the operational range of the SF-4 spectrophotometer by using filters with a fixed degree of color references. In this case it is possible to measure densities up to 5 units, which then permits the measurement of concentration of color centers nearing the level of 10^{19} cm^{-3} .

Conclusions

1. The magnitude of stored energy resulting from proton irradiation is a function of the type of the alkali halide crystals, i.e., the greater is the lattice energy of the crystal, the higher is the energy stored in it.
2. Within the investigated interval of the proton irradiation doses, a two-stage character of accumulation of the stored energy was established: a fast and a slow one. The fast stage of accumulation is related to F-centers, the slow one, to complex electron centers.

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